EFFICIENCY ENHANCEMENT OF EXCITONIC SOLAR CELLS THROUGH ACTIVE LAYER OPTIMIZATION

SHORT SYNOPSIS

Submitted in partial fulfillment of the requirement of the degree of

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Certificate

I hereby declare that the short synopsis **"Efficiency Enhancement of Excitonic Solar Cells Through Active Layer Optimization"** being submitted in partial fulfillment of the requirement for the degree of Doctor of Philosophy in Electronics and Communication Department under School of Engineering and Technology of Manav Rachna International Institute of Research & Studies, Faridabad during the academic year 2021-2022

It is a bona fide record of original work carried out under the supervision of Dr. Ashish Grover & Joint- Supervisor Dr. Pardeep Kumar and has not been submitted in full or part to any other university or institute for degree or diploma. Further, it is certified that short synopsis is plagiarism free and proof has been submitted.

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ABSTRACT

Amid escalating global warming concerns and surging crude oil prices, nations throughout the world have dramatically ramped up investments in renewable energy research and development, with a pronounced focus on solar energy. The renewable energy sector could undergo a significant transformation thanks to excitonic solar cells which are low cost, easy to manufacture, and lightweight. Excitonic solar cells have yet to match the efficiency of traditional solar cells, despite significant strides made in this area. As a result, there is an urgent need to produce high-efficiency excitonic solar cells. This research is to make more efficient excitonic solar cells by active layer optimization through a harmonious fusion of theoretical analysis and sophisticated modelling techniques.

The research focuses on improving device efficiency by exploring different donor acceptor materials in the active layer development of organic solar cells (OSCs) and also efficiency enhancements of perovskite solar cells (PSCs). Ultimately, this research aims to achieve performance enhancement, leverage the general advantages of excitonic solar cells, predict their performance characteristics, and pursue the overarching goal of cost reduction while enhancing their efficiency. This research contribution aligns with the broader mission of promoting the sustainable development of OSCs and PSCs within the renewable energy landscape.

Key words: Excitonic Solar Cells, Organic Photovoltaic Cells, Polymers, Perovskite Solar Cells, Acceptors, Power Conversion Efficiency.

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1. Introduction

The destiny of global energy is clearly a far more pressing issue that is being tackled by the scientific world. Indeed, our society could not function in the absence of energy. We cannot diagnose or help patients, nor can we filter water, drive automobiles, use computers, or study at night. Currently, fossil fuels constitute the majority of the world's energy consumption [1]. CO2 emissions, together with resource constraints and non-availability, are compelling reasons to oppose the use of fossil fuels [1]. Alternatives or renewable energy types must contribute to a growing proportion of the energy supply as time goes on. Renewable energy sources, such as solar thermal, wind, tide power, geothermal, and marine now make up a relatively small portion of the world's primary source of energy [2]. Solar energy is a fantastic solution to a sustainable and reliable energy source. "A photovoltaic (PV) or solar cell converts the energy of sunlight directly into the electricity by use of photovoltaic effect" [3]. Becquerrel was the first to find Photovoltaic effect [4]. Initially, Bell Labs Chapin et al. (1954) developed an inorganic solar cell with a 6% efficiency [5]. People began to fantasise about putting solar cells everywhere they could, but this was not feasible. The study advanced to explore various generations of solar cells, as depicted in Fig.1.

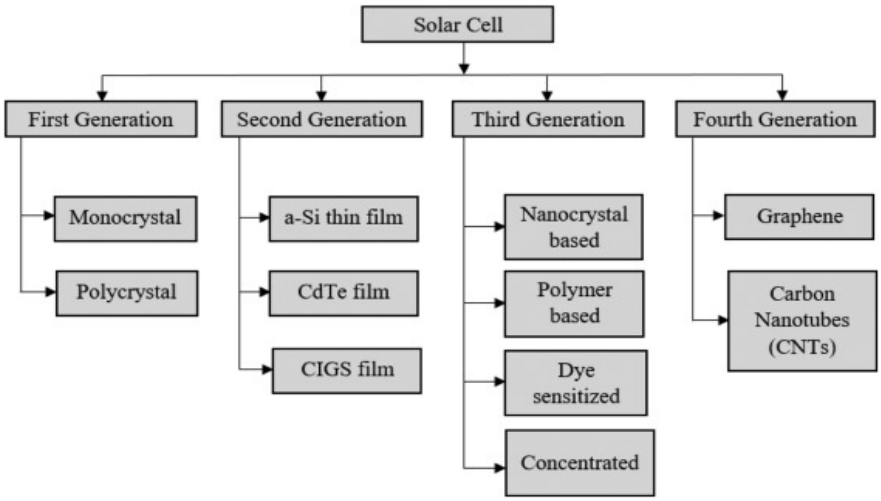

Fig. 1. Schematic representation of solar cell generations [6]

Crystalline silicon (Si) was used in the initial generation of solar cells. Si is the most prevalent material used in PV cells. Due to the high manufacturing costs, high material consumption, high embodied energy, lengthy energy payback period, and high production temperature required to develop a suitable material for solar cells, pure silicon must be

processed. Since pure silicon lacks the necessary characteristics required for solar cells, it must be processed extensively. It is a poor electrical conductor. Si has a 1.1 eV energy band gap. It is a nontoxic substance. For the abovementioned reasons, the second generation of solar cells comes into play. The thin-film solar cells are the second generation. Amorphous Si, CdTe thin film, and CIGS thin film are among the materials used. These solar cells address all of the issues raised by first-generation solar cells, but they have low stability. The third generation of solar cells eliminates all of these concerns. Organic solar cells (OSCs) and Perovskite solar cells (PSCs) are classified within the third generation. The polymer or plastic solar cells are another name for organic solar cells. The Nobel Prize in Chemistry was awarded to Alan J. Heeger, Alan MacDiarmid, and Hideki Shirakawa in 2000 for their discovery that polymers can function as semiconductors [7]. A new field of study was launched as a result of the realization that conjugated polymers can transmit electrons when doped with iodine and used to create solar cells. Krebs et al. (2008) classified the polymer solar cell unification problem into three issues: efficiency, stability, and processing [8] [9]. So researchers and scientists are focusing on these areas to improve. This research is primarily concerned with increasing the efficiency of OSC and PSC.

2. Literature Review

A thorough literature review was carried out and presented in a clear manner. According to the survey, the following trend of increase in research on the issue has emerged.

2.1 Organic Solar Cells: A Journey of Active Layer Development

In conformity with the National Renewable Energy Laboratory's (NREL) investigation and graphical depiction of the PCE of OSCs in 2023, it has been observed a substantial growth in the PCE, reaching up to 19% in recent years [10] . The first type of OSC with a single active/dynamic layer was demonstrated by Marks et al. (ITO-PPV-metal). However, single layer devices have PCE < 0.1% due to challenges in creating efficient excitons via dissociation and considerable charge carrier recombination [11]. Tang et al. invented bilayer heterojunction OSC with a donor (D) and acceptor (A) in 1986, obtaining a PCE ~ 1% [12]. It was recognised as a significant advancement in the province of OSCs. Yu et al. constructed an OSC with bulk heterojunction (BHJ) in 1995, in which a donor combined with an acceptor acted as the active layer [13]. This OSC structure increased the D/A interface while simultaneously considerable decreasing the diffusion distance for exciton

dissociation, leading to a boosted effectiveness of cells [13]. This OSC creation, which was marked as a triumph in OSCs, increased the efficiency of OSCs to above 18%, making it a potential photovoltaic technology [13] [14].

2.1.1 Donor (D) – Acceptor (A) Materials in Active Layer Development for OSCs

D/A materials are extremely significant in OSCs. There have been two research stages in the creation of acceptor materials. Prior to 2015, fullerene derivatives were the primary acceptors of OSCs polymers [15] . Lin et al. (2015) demonstrated increased absorption, higher mobility of electrons, and enhanced miscibility of D/A in a non fullerene acceptor (NFA), ITIC [16]. Since that day, NFAs have captured substantial recognition, and the performance of OSCs using NFA has improved dramatically over the years.

2.1.1.1 Fullerene and Non Fullerene: Acceptor Evolution in Active Layer

Mikroyannidis et al. developed an OSC using the active layers P3HT:F and P3HT:PC61BM, yielding efficiencies of 4.23% and 2.93%, respectively [15] [17]. Thermal annealing increased the efficiency of the P3HT:F film to 5.25% [15]. Wienk et al. produced an OSC in 2003 by synthesising PC71BM, a stronger fullerene ring acceptor than PC61BM, and combining it with MDMO-PPV to yield PCE at roughly 3% [18]. Troshin et al. created an P3HT:PC71BM based OSC and achieved a PCE of 4.1% [19]. Yu et al. exploited narrow band gap donor materials like PTB7 to develop a PTB7:PC71BM based OSC with a PCE of 7.4% [14]. In 2014, Liu et al. created an OSC with PffBT4T-2OD: PC71BM as donor and acceptor materials, advancing OSCs to a new level of PCE of more than 10% [20]. Albeit fullerene acceptors have received a lot of engrossment in the domain of OSCs, they have some serious weaknesses, such as: (1) dim near-infrared (NIR) absorption and visible spectral ranges, (2) limited tunability of energy levels and chemical structure, (3) morphological instability, and (4) pricey production outlay. These disadvantages have prompted researchers to investigate NFAs, including small molecules and polymers, for enhanced efficiency in OSCs [21] [22] [15]. Compared to fullerene acceptors, NFAs have notable benefits in optical absorptivity, frontier orbital energy bandgap, levels, and tunability [23] [15]. In the preceding years, the NFA-based OSCs has made tremendous development, and the efficacy record of OSCs with NFAs has been regularly renewed. Zhan et al. created and reported an OSC utilising a novel acceptor material, ITIC, achieving a PCE of 6.8% in 2015 [24]. Additionally, Zhao et al. reported an OSC with an efficiency of 11.21% with ITIC

as an acceptor [14]. Similarly, Xu et al. stated an efficacy of 12.80% by creating an OSC with PBDTS-TDZ (donor) and ITIC (acceptor) [25]. Later, Yuan et al. discovered a novel class of NFA, "Y6, in which dithienothiophen[3,2-b]-pyrrolobenzothiadiazole (TPBT), a ladder-type electron-deficient-core-based central fused ring", was utilised. In addition, the investigation found an OSC with a PCE of 15.7% [26]. The optical bandgap of the Y6 is 1.33 eV [26]. The PCE of NFA-based OSCs has been significantly improved thanks to the assistance of Y6. Subsequently, Tran et al. created a PBDB-T-2F:Y6-based OSC with a PCE of 17.1% [27]. Liu et al. then reported an OSC with D18:Y6 as the active layer, and the cell had the highest PCE of 18.22% [28]. Many people were interested in Y6 and its derivatives because of its exceptional photovoltaic performance. Li et al. recently reported on a class of NFAs, L8-BO [29] [32]. In the study, an OSC with PM6:L8-BO as the active layer was built, and a phenomenal PCE of 18.32% was attained [29]. Subsequently, using PM6: L8-BO as the active layer, Meng et al. reported an OSC with a PCE of 18.77% [30]. OSCs including small molecule NFAs like as ITIC, Y6, and L8-BO have demonstrated outstanding PCEs, making them a potential platform [15]. Then, another fascinating technique for generating bulk heterojunction OSCs made entirely of polymeric semiconductors, known as "all-polymer solar cells", is explained further in detail [15]. To date, the most common monomers used to construct polymeric NFAs in all-polymer solar cells include B←N-bridged bipyridine (BN-Py), perylene diimide (PDI), bithiophene imide (BTI), and naphthalene diimide (NDI) [15]. Solar cells incorporating these materials have also demonstrated remarkable PCEs [15].

2.1.1.2 Evolution of Donor Materials in the Active Layer of OSCs

In addition to the existence of effective acceptors, donors play an important role in the progression of organic solar cells (OSCs) [15]. Conjugated poly (phenylene vinylene) (PPV) and its derivatives sparked considerable interest in the early phases of bulk heterojunction organic solar cells (BHJ-OSCs), owing to their conductive and photoluminescent properties [15]. "On the basis of optical bandgaps (Eg opt), donor materials can be divided into three major types: wide bandgap (WBG, Eg opt > 1.8 eV), medium bandgap (MBG, 1.6 eV <Eg opt≤ 1.8 eV), and low bandgap (LBG, Eg opt ≥1.6 eV)" [31][15]. Mhlbacher et al. (2006) used LBG PCPDTBT as donor material to create an OSC with a PCE of 3.2% [32] [33]. Building upon this progress, Chen et al. (2009)

employed PBDTTT-CF as the donor achieving a significantly improved PCE of 7.73% [34]. Shortly after, Lu et al. synthesized and constructed an OSC based on PTB employing LBG polymers, achieving a claimed PCE of 5.90% [35]. He et al. successfully developed an OSC employing PTB7 in 2019, yielding a higher reported PCE of 9.214% [36] . The Hous et al. then synthesized PBDB-T and used it in OSCs with fullerene acceptors, yielding a PCE of 6.67% [37]. Furthermore, when PBDB-T was employed as the donor material in an OSC with a NFA, a PCE of 11.21% was obtained [38]. Yuan et al. followed in 2019, using PBDB-T as the donor material and reporting an excellent efficiency of 13.42% [39]. Later, PBDB-T-2F (PM6) was introduced. Later, the introduction of PBDB-T-2F (PM6) and PBDB-T-2Cl came into play. In 2019, Yuan et al. created an OSC using PM6 as the donor, resulting in a remarkable PCE of 15.7% [26]. Using the extraordinary features of the revolutionary donor material, D16, Xiong et al. (2019) showed an excellent PCE of 16.72% [40]. Later, Liu's group developed D18 and reported a PCE of 18.22% [28].

2.2 Perovskite Solar Cells: An Ongoing Journey of Progress

In conformity with the NREL research study chart on the PCE of PSCs in 2023, it has seen a substantial growth in the PCE, reaching up to 25.73% in recent years [41]. Scientists have been studying organic-inorganic halide perovskites for over a decade, with the roots of perovskite solar cells (PSCs) dating back to 1839, when German physicist Gustav Rose unearthed an entirely novel mineral based on CaTiO3 while visiting Russia and named it "perovskite" after Russian mineralogist Lev von Perovski [42]. In 2006, Miyasaka et al. documented the achievement of a PCE of 2.2% in CH3NH3PbBr3 cells, and in 2009, reported further efforts that increased the PCE to 3.8% [43] [44]. However, all of the devices exhibited instability. Park et al. were able to achieve a PCE of 6.5% at a later time by creating the perovskite sensitizing material on a TiO2 film. However, this result is consistent with their study published in 2011 [45]. According to the study by Kim et al. (2012), a solid-state PSC with a PCE of 9.7% [46]. This innovation accelerated research into hybrid organic-inorganic PSCs. The PCE of PSC-based devices demonstrated a quick and substantial increase, progressing from 15% to a remarkable 25.73%[41] [47] [48] [49] [50] [51].

3. Description of Broad Area

3.1 Working of Organic Solar Cells (OSC)

An OSC creates electricity and is represented in the several major phases of the conversion process, which are outlined.

1. Photon absorption and creation of excitons: A photon incident on a semiconductor with an energy greater than the band gap excites an electron to an unoccupied state above the band gap, resulting in the formation of an electron-hole pair (e-h) [52] .
2. Exciton diffusion and charge separation: The e-h pair is then separated along a built-in gradient in the electrochemical potential [52].
3. Charge transport: Finally, after being sent to operate in an external circuit, the electron and hole are collected in opposing electrodes and recombine [52].

3.2 Build-up of OSC

OSCs are commonly created by dissolving polymers in organic solvents and printing or coating them onto a substrate [52] [53]. To build a photovoltaic stack, the materials are arranged in layers in a predefined manner. The following materials are required for the photovoltaic stack:

1. a central or core active (light-absorbing) layer: A donor material and an acceptor material make up the active or dynamic layer. Polymers serve as donors in the OSC. The combination of monomers results in polymers. In OSCs, thiophene acts as the backbone [54]. Side chains can be either alkyl chains or esters to make polymers soluble. For polymers, it is crucial that they be conjugated, or situated between single and double bonds. Pi electrons can be found in the double bond. Visible light is absorbed by the pi bonds, which cause it to shift from HOMO to LUMO. This transfer is known as Pi to Pi* [54].
2. Transport Layers: Due to an appropriate arrangement of the energy levels, the materials that make up the transport layers can predominantly move either electrons or holes. Hole transport layers (HTL) include PEDOT:PSS, P3HT, MoOx, whereas electron transport layers (ETL) include ZnO, LiF, Ca, TiOx, and others [54].
3. Electrodes: There are basically two electrodes, and one is transparent to allow enough light into the solar cell. Example: ITO, FTO, etc. [54].

4. Substrate: Glass and polymers are the two substrate types utilised in polymer solar cells. During the fabrication of OSC, substrates are employed to support the layered photovoltaic stack [54].

Normal and inverted geometries (Fig.2) are available in OSC. In conventional OSC, the substrate/anode/ETL/active layer/HTL/cathode is the order, however in inverted OSC, the substrate/Cathode/HTL/active layer/ETL/anode is the order.

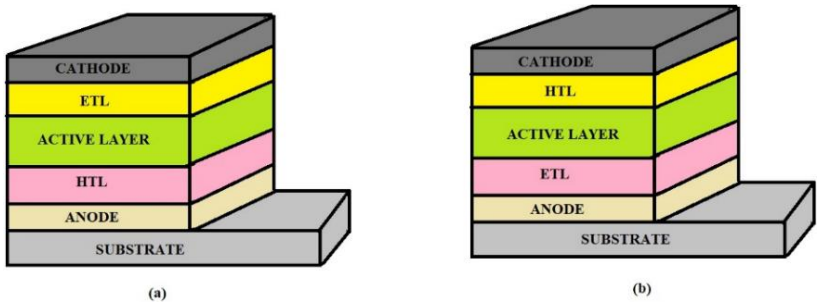

Fig. 2. (a) Normal (Conventional) OSC, (b) Inverted OSC

3.3 Working of Perovskite Solar Cell (PSC)

Methylammonium lead iodide (MAPbI3) is the prevailing choice of material for the active layer in PSCs [55]. This layer sits between an ETL and an HTL [55]. The ETL is commonly constructed of mesoporous titanium dioxide (TiO2) but can alternatively be planar TiO2 [55]. The transparent front electrode in a common device configuration is a glass substrate coated with fluorine-doped tin oxide (FTO), while the back electrode is a layer of gold formed by thermal evaporation [55].

3.4 PSC Build UP

Fig. 3 depicts the crystal structure of organic-inorganic perovskite, which has the typical chemical formula ABX3[55].

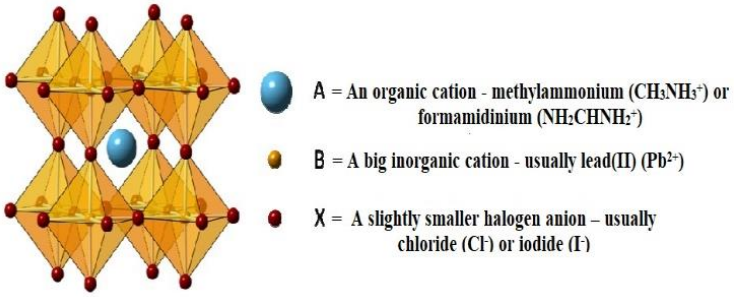
A = An organic cation - methylammonium ($CH_3NH_3^+$) or formamidinium ($NH_2CHNH_2^+$)
B = A big inorganic cation - usually lead(II) (Pb^{2+})
X = A slightly smaller halogen anion – usually chloride (Cl^-) or iodide (I^-)

Fig. 3. Perovskite Structure [55]

PSCs are made up of a core perovskite layer sandwiched between an ETL and a HTL. If the structure is arranged in FTO / ETL / perovskite / HTL / electrode, then it is called a n-i-p (conventional) structure and ITO/HTL/perovskite/ETL/electrode is called an p-i-n (inverted) structure [56]. PSCs can also be classified on the basis of materials used [56]. The continued study of this emerging solar technology has explored several pathways by utilising a diverse range of materials [56]. The integration of mesoporous materials was the first step, and over time, planar structured substances, carbon-based compounds, quantum dot materials, and other materials were incorporated to attain outstanding levels of performance [56]. The following sections explain the various types of PSCs.

1. Planar PSC - A sandwich-like structure composed of p-type and n-type materials, formed by a low-temperature method, is frequently used in planar PSCs, both with and without an ETL or a HTL [56].
2. Mesoporous PSCs - Mesoporous PSCs predominantly adhere to the conventional (n-i-p) sandwich construction that includes a mesoporous layer. In the beginning, devices featuring mesoporous layers were constructed using mesoporous TiO2. Following that, substantial progress was achieved by including a mesoporous spacer layer, a HTL, and a carbon-based electrode layer [56].
3. Although Mesoporous and Planar structures share the task of charge separation, they differ in how charge transport and recombine [57]. The mesoscopic structure is found to be less effective than the planar structure due to a higher rate of charge recombination, even though they have the same speed of charge movement [57]. This recombination problem reduces the efficiency of the mesoscopic structure, distinguishing it from the more efficient planar form [57] .
4. Carbon Based PSC- Carbon's extraordinary properties stem from its diverse bonding structures and allotropes [56]. Carbon has made major contributions to several scientific and technological sectors [56]. Its environmentally favourable characteristics and cost-effective procedures make it a great candidate for PSCs. Carbon has been used as a transparent conductive layer, ETL, HTL and even as a counter electrode layer in this respect [56].

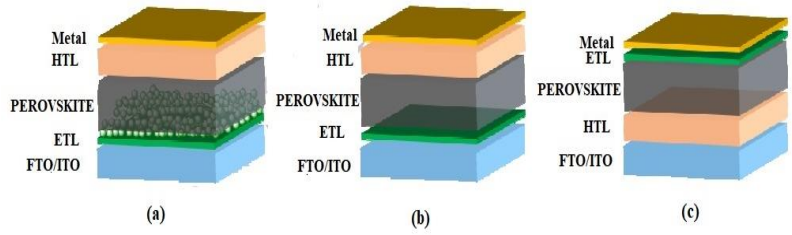

Fig. 4. (a) Mesoporous PSC, (b) Planar PSC and (c) Inverted PSC [55]

3.5 V-I characteristics

When selecting a solar cell for a certain project, it is critical to understand the solar cell's rating. These parameters indicate how well a solar cell converts light into electrical energy.

1. Open circuit voltage (V_{OC}): This is present in the circuit when the cell's two terminals are unconnected or when $Rl = \infty$ [58].
2. Short circuit current density: J_{SC} stands for Short circuit current density. This is observed in circuit when cell's two terminals are connected or when $Rl = 0$ [58].
3. Pm, Im and Vm: The Pm in the figure shows the maximum power point of a solar cell. The current at maximum power point, indicated by "Im," is the current at maximum power. The voltage at the maximum power point, abbreviated Vm, is the voltage at which maximum power occurs [54].

   The relationship between the above three is as follows: $Pm = Im \times Vm$ (1)
4. Fill Factor: The ratio of Pm to (Isc x Voc) is used to calculate the fill factor.

$$\mathrm{FF} = \frac{\mathrm{Pm}}{\mathrm{Isc} \times \mathrm{Voc}} \quad (2)$$

5. Power conversion efficiency (PCE): It is defined as the ratio of output power to input power, which can alternatively be expressed as the product of FF, I_{SC}, and V_{OC} [54].

$$\mathrm{PCE} = \frac{\mathrm{Pm}}{\mathrm{Pout}} \text{ or } \mathrm{PCE} = \mathrm{FF} \times \mathrm{Isc} \times \mathrm{Voc} \quad (3)$$

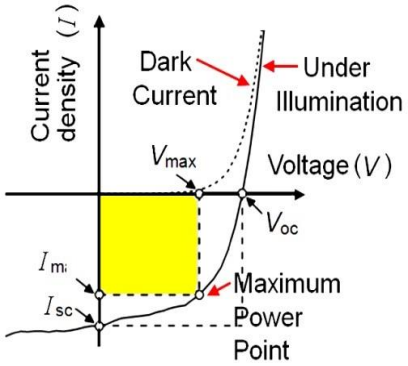

Fig. 5. V-I Characteristics of OSC and PSC [59]

4. Objectives of Research

The following research objectives are identified to accomplish the aforementioned goal in an elegant manner.

1. To review organic solar cells (OSCs) with different active layer materials and perovskite solar cells (PSCs) for analysing efficiency improvements.
2. To perform Device Modelling and Analysis of OSCs and PSCs.
3. To perform comparative analysis between the performance of OSCs using different Active layer materials.
4. To design and evaluate for analyzing efficiency of Excitonic cells using different softwares.

5. Methodology to be Adopted

The research will be conducted in accordance with the subsequent procedure:

1. Review the existing organic solar cells (OSCs) with different active layer materials and perovskite solar cells (PSCs) for analysing efficiency improvements.
2. Designing and Modelling OSCs and PSCs in a computational environment to accurately represent their behavior and performance.

Parameter sweep: This technique entails systematically changing one or more parameters in the simulation and analysing the influence on the performance of the OSCs and PSCs. This can be used to uncover design aspects that have a large influence on cell efficiency and to optimize device architecture or material selection.

The parameters mentioned above will include the following:

Different active layer materials, thickness, bandgap, carrier mobility, absorption coefficients, exciton binding energy, interfacial layer thickness, electrode materials, device geometry, device dimensions, structure, etc.

3. Comparative analysis entails evaluating the simulated performance of several OSC and PSC designs or materials and analyzing the advantages and disadvantages of each choice. This may be used to assess the possibilities of various methods and to advise material or architecture decision.

4. Designing and evaluating using different software to assess efficiency improvements in Excitonic Cells.

6. Expected Outcome of the Research

This Research study might produce beneficial results.

1. General advantages of OSC and PSC can be implemented such as: Low weight, flexibility, Semitransparency, seamless integration, new market prospects, significantly lower manufacturing costs, continuous production via advanced printing tools, short energy payback times and minimal environmental footprints [60].
2. Device architecture optimization or material selection: Simulation outcomes can suggest various design elements that can improve the efficiency of OSCs and PSCs such as modifying the thickness and composition of each layer in the device architecture or optimizing the geometry of the electrodes.
3. Predicted performance characteristics: The simulation results provide information about the expected performance of OSCs and PSCs under various scenarios, such as maximum PCE, FF, or Voc.
4. The study aims to mitigate the expenses associated with producing OSCs and PSCs while also improving or retaining their efficacy.

7. Proposed Time Frame

Time Frame	Feb 22 – Aug 22			Sep 22 - Feb 23			Mar 23- Aug 23			Sep 23- Feb24			Mar 24 - Aug 24			Sep 24- Feb 25			May 25		
Months	2	4	6	8	10	12	14	16	18	20	22	24	26	28	30	32	34	36	37	38	39
Course Work																					
Extensive Literature Survey																					
Problem Statement																					
Modelling and analysis of OSCs and PSCs.																					
Comparative study between the performance of OSCs using different Active layer materials.																					
Designing and evaluating efficiency of Excitonic cells using different softwares.																					
Publication within the context of the study.																					
Full-scale report creation and submission.																					

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